

# Possible Pairing Mechanisms of PuCoGa<sub>5</sub> Superconductor

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We examine possible pairing mechanisms of superconductivity in PuCoGa<sub>5</sub> based on spin-fluctuations or phonons as mediating bosons. We consider experimental data of specific heat C(T) and resistivity  $\rho(T)$  as input to determine a consistent scattering boson with the superconducting transition temperature of 18.5 K in PuCoGa<sub>5</sub>. Irrespective to the type of boson, the characteristic boson frequency is found to be  $\sim 150$  K from the resistivity fitting. The spin fluctuation model is most consistent with the experimental resistivity, successfully explaining the anomalous temperature dependence ( $\sim \frac{T^2}{150K+T}$ ) at low temperatures as well as the saturation behavior at high temperatures. Assuming that the pairing state is non s-wave, the large residual resistivity  $\rho_{imp} \sim 20\mu\Omega cm \sim 120$  K suggests that an ideally pure sample of PuCoGa<sub>5</sub> would have a maximum T<sub>c</sub> of 39 K.

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## I. INTRODUCTION

Recently, superconductivity was found in PuCoGa<sub>5</sub> at the amazingly high transition temperature (T<sub>c</sub>) of 18.5 K<sup>1</sup>. Considering the fact that the highest T<sub>c</sub> of f-electron based compounds<sup>2</sup> was  $\sim 2$  K, the T<sub>c</sub> of 18.5 K is an order of magnitude larger value than the previous highest T<sub>c</sub> in f-electron based superconducting compounds. Therefore, the understanding the origin of this 18.5 K transition temperature in PuCoGa<sub>5</sub> should not only provide important information on the puzzling behavior of f-electrons but also shed light on the origin of the high transition temperature in cuprate superconductors.

Let us briefly review the experimental data known about PuCoGa<sub>5</sub>. First, from the specific heat jump at T<sub>c</sub>, the Sommerfeld coefficient is  $\gamma_{normal} \sim 77$  mJ/K<sup>2</sup>mol. Second, from the T<sup>3</sup> phonon contribution in C(T), the typical value of the phonon frequency scale ( $\Theta_D$ ) is estimated to be 240 K. Third, the resistivity  $\rho(T)$  shows a typical S-shape behavior in its temperature dependence, which is often observed in spin fluctuating heavy fermion compounds such as UPt<sup>3</sup> [3]. Another important piece of information from  $\rho(T)$  is its magnitude; the value  $\rho(T = 300K) \sim 250\mu\Omega$  cm itself indicates strong scattering of the conduction electrons (an order of magnitude larger than the values of CeMIn<sub>5</sub> (M=Co,Ir,Rh) superconductors having T<sub>c</sub> of  $\sim 2$  K<sup>2,4</sup>). Forth, puzzling is the data of the uniform susceptibility  $\chi(T)$ ; it shows an almost exact Curie-Weiss temperature dependence of  $\chi(T) \sim 1/(T + T_\theta)$  with  $T_\theta = 2$  K. This indicates that there are almost free local moments in the temperature range of 18.5 to 300 K with an effective local moment magnitude (0.68  $\mu_B$ ) close to the value of the local moment of free Pu<sup>3+</sup> (0.84  $\mu_B$ )<sup>5</sup>. We believe that these local moments responsible for the observed  $\chi(T)$  are not coupled (or negligibly weakly coupled) to the conduction electrons and play no significant role in

the transport properties as well as in the superconducting pairing. This does not mean that there are no interesting spin fluctuations – their contribution to  $\chi(T)$  might be much smaller than the local moment contribution or more probably their temperature dependence is not pronounced. If this is the case, the Curie-like  $\chi(T)$  should continue to exist below the transition temperature and should be observable if the diamagnetic part of  $\chi(T)$  is subtracted or suppressed below T<sub>c</sub>.

As a possible pairing mechanism and pairing symmetry, we do not have much decisive data except estimates of a few energy scales. First of all, conventional phonon mediated pairing seems not unreasonable but only barely possible with values of  $\Theta_D = 240$  K, dimensionless coupling constant  $\lambda = 0.5 \sim 1.0$ , and a typical value for the Coulomb pseudopotential  $\mu^* = 0.1$ . The Allen-Dynes' T<sub>c</sub> formula<sup>6</sup>  $T_c = \frac{\omega_{ph}}{1.20} \exp[-\frac{1.04(1+\lambda)}{(\lambda - \mu^*(1+0.62\lambda))}]$  provides T<sub>c</sub>=16.7 K for  $\lambda = 1$  and T<sub>c</sub>=2.9 K for  $\lambda = 0.5$ , respectively. On the other hand, although there is not yet direct experimental evidence, the existence of Pu f-orbitals participating in Fermi level crossing band(s)<sup>7</sup> and the isostructure CeMIn<sub>5</sub> (M=Co,Rh,Ir) compounds let us suspect the important role of spin-fluctuations to explain the normal state transport properties as well as the pairing mechanism in PuCoGa<sub>5</sub>.

In this paper, we examine two possible bosonic scattering mechanisms, namely, phonons and spin-fluctuations, to consistently understand the available experimental data mentioned above. Our strategy is the following. Assuming each bosonic scattering, we try to fit the dc-resistivity data  $\rho(T)$  for its temperature dependence as well as its magnitude. From this fitting procedure, we extract the magnitude of the dimensionless coupling constant  $\lambda$  and the typical energy scale of the corresponding boson. From these two numbers, we then can estimate T<sub>c</sub> using McMillan's formula.

## II. FORMALISM

We calculate the conductivity using the Kubo formula as follows.

$$\sigma(T) = \frac{\hbar e^2}{3} \sum_k v^2(k) \int \frac{d\omega}{4\pi T} A^2(\vec{k}, \omega) \left[ \frac{1}{\cosh^2[\omega/2T]} \right] \quad (1)$$

where  $A(\vec{k}, \omega) = 2\text{Im}G_R(\vec{k}, \omega)$  is the one particle spectral density of quasiparticle in the conduction band and the retarded Green function of the quasiparticle is defined as  $G_R(\vec{k}, \omega) = \frac{1}{\omega - \epsilon_p - \Sigma(\vec{k}, \omega)}$ . All scattering information is included in the self-energy  $\Sigma(\vec{k}, \omega)$ . Within the Born approximation, the self-energy is calculated as

$$\Sigma(\vec{k}, \omega_n) = g^2 T \sum_{\Omega_n, q} \int d\omega' \frac{B(q, \omega')}{i\Omega_n - \omega'} \frac{1}{i\omega_n + i\Omega_n - \epsilon_{k+q}}, \quad (2)$$

where  $B(q, \omega)$  is the spectral density of a bosonic propagator  $D(q, \omega) = \int \frac{d\omega'}{2\pi} \frac{B(q, \omega')}{i\Omega_n - \omega'}$  and  $g$  is the electron-boson coupling constant. After a Matsubara frequency ( $\Omega_n$ ) summation, the imaginary part of  $\Sigma_R(\vec{k}, \omega + i\eta)$  is written as

$$\begin{aligned} \text{Im}\Sigma_R(T, \omega + i\eta) &= g^2 N(0) \int \frac{d\omega'}{2\pi} \quad (3) \\ &\times \sum_q \pi B(q, \omega') [n(\omega') + f(\omega + \omega')] \end{aligned}$$

where  $n(\omega)$  and  $f(\omega)$  are the Boson and Fermion distribution functions, respectively.  $N(0)$  is the density of states per spin at the Fermi level. Plugging the self-energy Eq.(2) into Eq.(1) and summing  $\sum_k$ , Eq.(1) gives

$$\sigma(T) = \frac{\hbar e^2}{3} N(0) \langle v^2 \rangle_{FS} \int \frac{d\omega}{4T} \left[ \frac{1}{\cosh^2[\omega/2T]} \right] \frac{1}{\text{Im}\Sigma_R(T, \omega)} \quad (4)$$

A few remarks are in order for the above equations. First, the vertex correction is ignored. The justification is that when the scattering is local in space – technically meaning that the self-energy is momentum independent – the current vertex is not renormalized. This is consistent with the local approximation in calculating Eq.(2) and Eq.(3); consistent with this, we also neglect the momentum dependence of the coupling  $g$  implying every quasiparticle is equally scattered by the assumed boson. Second, the self-energy is calculated only in the Born approximation. Third, the Fermi surface (FS) anisotropy is neglected, resulting the factor  $\frac{1}{3}$  and the FS averaged Fermi velocity squared  $\langle v^2 \rangle_{FS}$ . With all these approximations, we should take the temperature power law of the calculated resistivity at low temperatures with reservation. Otherwise it induces an error of order  $O(1)$  for

the overall magnitude. Finally,  $N(0)$  and  $\langle v^2 \rangle_{FS}$  are the values before they are renormalized by the bosonic scattering and, therefore, are difficult to be estimated from experiments. We rewrite Eq.(4) as follows.

$$\sigma(T) = \frac{\hbar e^2}{3} Z \tilde{N}(0) \langle \tilde{v}^2 \rangle_{FS} \int \frac{d\omega}{4T} \left[ \frac{1}{\cosh^2[\omega/2T]} \right] \frac{1}{\text{Im}\Sigma_R(T, \omega)} \quad (5)$$

In the above equation,  $\tilde{N}(0)$  and  $\tilde{v}$  are the quantities renormalized by the bosonic scattering and  $Z$  is the wave function renormalization parameter  $Z = 1 + \partial \text{Re}\Sigma(\omega)/\partial\omega$ . The above expression is very useful for our purpose. The renormalized quantities  $\tilde{N}(0)$  and  $\tilde{v}$  can be obtained from experiments. Furthermore, although Eq.(5) has an explicit dependence on  $Z$ , the implicit dependence of  $\text{Im}\Sigma(T, \omega)$  on  $Z$  makes Eq.(5) a slowly varying function of  $Z$ . The reason is because the real and imaginary parts of  $\Sigma(T, \omega)$  are related by the Kramer-Kronig relation, so that  $\partial \text{Re}\Sigma(T, \omega)/\partial\omega = Z(T) - 1$  resulting  $\text{Im}\Sigma(T, \omega) \sim [Z(T) - 1]$ .

## III. RESULTS

### A. Spin fluctuations

We choose the mean field type spin relaxational mode of  $B(q, \omega) = \frac{C\omega}{[I(T) + b(\vec{q} - \vec{Q})^2]^2 + [\frac{\omega}{\Gamma}]^2}$ , where  $\Gamma$  is the magnetic Fermi energy<sup>8</sup>,  $I(T) = I_0 + aT$  is the parameter controlling the distance from a magnetic quantum critical point, and  $\vec{Q}$  is a typical wave vector of the magnetic ordering.  $\Gamma \cdot I(T) = \omega_{sf}(T)$  defines the characteristic energy scale of the fluctuations.  $b$  describes the dispersion of the collective mode of  $D(q, \omega)$ . The magnetic ordering vector  $\vec{Q}$  can be in two dimensions or in three dimensions depending on the nature of the incipient magnetic order. This dimensionality of magnetic ordering would affect the power law of the resistivity at low temperatures. We assume 2-D in our calculations, reflecting the band calculations<sup>7</sup>. The overall magnitude  $C$  of  $B(q, \omega)$  is combined with  $g^2 N(0)$  in Eq.(3) to determine the overall magnitude of  $\Sigma(T, \omega)$ . This overall magnitude is determined once  $(Z(T) - 1)$  is fixed. Therefore, we do not need to determine separate values of  $g^2$ ,  $N(0)$ , and  $C$ . Most importantly,  $I_0$  determines the low temperature  $T^\alpha$  region of the resistivity before the inflection point;  $\alpha \sim 4/3$  was extracted from experiments<sup>1</sup> for  $T_c < T < 50\text{K}$ , but a dimensional counting for the spin fluctuations model in 2-D gives  $\rho(T) \sim \frac{T^2}{I_0 + aT}$  at low temperatures. The temperature variation of  $aT$  in  $I(T)$  also controls the high temperature saturation behavior of  $\rho(T)$ , since increasing value of  $I(T)$  for larger temperatures reduces the scattering rate.

Fig.(1) shows the typical results from Eq.(5) with  $Z$  values varying from 2 to 8. To be quantitative, the experimental values of  $\tilde{N}(0)$  ( $\gamma = 77\text{mJ/K}^2\text{mol}$ ) and

$\tilde{v} (= 8.8 \times 10^6 \text{ cm/sec})^5$  are used. Given these two experimental input values, there is no free parameter to adjust the overall magnitude of the resistivity. The detailed temperature dependence of  $\rho(T)$  is controlled by  $I(T)$ .  $\Gamma \cdot I_0 = \omega_{sf} = 150 \text{ K}$ ,  $a = 1$  are chosen for illustration. As seen in Fig.(1), with increasing  $Z$ , the sensitivity of  $\rho(T)$  to  $Z$  becomes weaker, and for all values of  $Z$  the calculated  $\rho(T)$  is smaller than the experimental one by a factor of 3 to 4 (see Fig.2). In order to fit the experimental  $\rho_{exp}(T)$ , we tune the value of  $\tilde{v}$ , which is a rather rough estimate from the critical magnetic fields ( $H_{c2}(0)$ ) in superconducting state of PuCoGa<sub>5</sub>, while the value of  $\gamma_{exp} = 77 \text{ mJ/K}^2 \text{ mol}$  is more reliable. We also chose to use  $Z = 4.6$  which is the ratio of  $\gamma_{exp}/\gamma_{band}$ <sup>7</sup>. The result is shown in Fig.(2) in comparison with the experimental  $\rho_{exp}(T)$ . Input parameters are  $\omega_{sf} = 150 \text{ K}$ ,  $a=1$ ,  $\gamma = 77 \text{ mJ/K}^2 \text{ mol}$ , and  $\tilde{v}_{exp} = 4.78 \times 10^6 \text{ cm/sec}$ . The residual resistivity  $\rho_{imp} = 15 \mu\Omega \text{ cm}$  is added to the theoretical result (The better fitting of the low temperature part only would give  $\rho_{imp} = 19 \mu\Omega$ . See the inset of Fig.(2).) The overall fitting is satisfactory from low to high temperatures. In particular, the saturation behavior at high temperatures is well reproduced with the temperature dependent  $I(T)$ . This is an expected behavior since increasing temperature should shorten the magnetic correlation length such as  $\xi^{-2}(T) \sim (I_0 + aT)$ . Some discrepancy between the theoretical result and the experimental one beyond 250 K (Fig.2) would be due to the failure of the simple relation  $\xi^{-2}(T) \sim (I_0 + aT)$  at such high temperatures. To obtain better fitting in the low temperature region, one needs to allow a modification of the high temperature part. The result is shown in the inset. It indeed reproduces the experimental observation  $\rho(T) \sim T^{4/3}$  [1] for  $T_c < T < 50 \text{ K}$ , while the correct theoretical form of  $\rho(T)$  at low temperatures is

$$\sim \frac{T^2}{150K+T}.$$

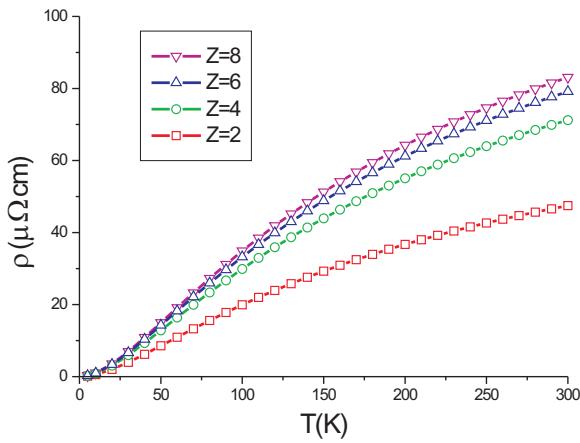


FIG. 1. Theoretical calculations of resistivity  $\rho(T)$  for varying  $Z$  values ( $Z = 2, 4, 6, 8$ ). Input parameters are  $\omega_{sf} = 150 \text{ K}$ ,  $a = 1$ ,  $\gamma = 77 \text{ mJ/K}^2 \text{ mol}$ , and  $\tilde{v}_{exp} = 8.8 \times 10^6 \text{ cm/sec}$ .

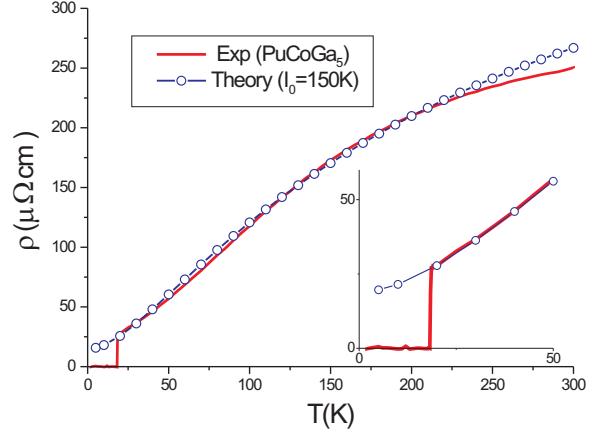


FIG. 2. Theoretical calculation with  $Z = 4.6$  (open blue circles) and the experimental resistivity  $\rho(T)$ <sup>1</sup> (red solid line). Input parameters are  $\omega_{sf} = 150 \text{ K}$ ,  $a = 1$ ,  $\gamma = 77 \text{ mJ/K}^2 \text{ mol}$ , and  $\tilde{v}_{exp} = 4.78 \times 10^6 \text{ cm/sec}$  and  $\rho_{imp} = 15 \mu\Omega \text{ cm}$  is added. Inset: closeup view of low temperature region. For better fitting,  $\tilde{v}_{exp} = 5.28 \times 10^6 \text{ cm/sec}$  and  $\rho_{imp} = 19 \mu\Omega \text{ cm}$  are used

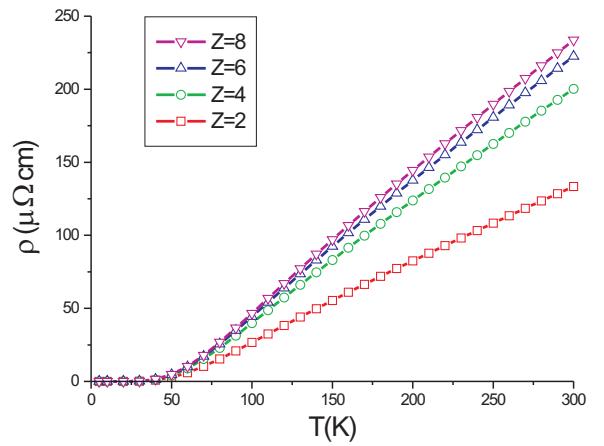


FIG. 3. Theoretical calculations of resistivity  $\rho(T)$  with Einstein phonon with  $\theta_D = 240 \text{ K}$  for varying  $Z$  values ( $Z = 2, 4, 6, 8$ ). Input parameters are  $\gamma = 77 \text{ mJ/K}^2 \text{ mol}$ , and  $\tilde{v}_{exp} = 8.8 \times 10^6 \text{ cm/sec}$ .

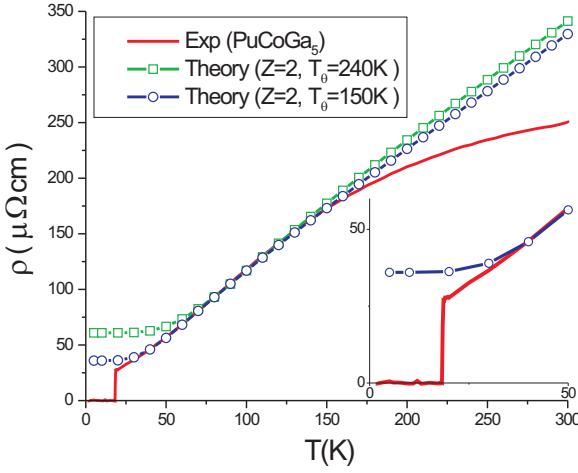


FIG. 4. Theoretical calculations of resistivity  $\rho(T)$  for  $Z = 2$  with two different Einstein phonon frequencies:  $\theta_D = 240\text{K}$ ,  $\gamma = 77\text{mJ}/\text{K}^2\text{mol}$ , and  $\tilde{v}_{exp} = 6.1 \times 10^6\text{cm/sec}$  (open green squares);  $\theta_D = 150\text{K}$ , and  $\tilde{v}_{exp} = 5.6 \times 10^6\text{cm/sec}$  (open blue circles). The experimental resistivity  $\rho(T)^1$  is red sold line. Inset: closeup view of low temperature region.

## B. Phonons

Fig.3 shows the resistivity calculated with an Einstein phonon  $B(q, \omega) \sim \delta(\omega - \theta_D)$  with  $\theta_D = 240\text{K}$ , which is the value obtained from the specific heat  $C(T)$  above  $T_c^1$ , for varying values of  $Z$  from 2 to 8. The input values are  $\gamma = 77\text{mJ}/\text{K}^2\text{mol}$  and  $\tilde{v}(= 8.8 \times 10^6\text{cm/sec})$ . As in the spin-fluctuations case (Fig.1), with increasing  $Z$  values the sensitivity of  $\rho(T)$  to  $Z$  becomes weaker, and the overall magnitude of  $\rho_{theor}(T)$  is smaller than  $\rho_{exp}(T)$  by the factor of 2 to 3 (compared the values at  $T=100\text{K}$ ). In Fig.4, we show the tuned theoretical results of  $\rho_{theor}(T)$  in comparison with the experimental  $\rho_{exp}(T)$ . As before, in order to increase the overall magnitude of  $\rho_{theor}(T)$ , we tune the renormalized Fermi velocity  $\tilde{v}$ .

In Fig.4, we see that the theoretical resistivity  $\rho_{theor}(T)$  with  $\theta_D = 240\text{K}$  (green open squares) has a higher power law region over a broader range of low temperatures relative to the experimental data. For overall fitting of the data, we add a large impurity resistivity  $\rho_{imp} = 61\mu\Omega\text{ cm}$ , which makes the low temperature part in clear disagreement with experiment. To fit the low temperature region better, we need to reduce  $\theta_D$ . The blue open circles are the result with  $\theta_D = 150\text{K}$ . This happens to be the same bosonic energy scale as used in the spin-fluctuation model fitting. It means that the resistivity data reveal a characteristic energy scale of the scattering boson to be  $\sim 150\text{ K}$ , irrespective of the origin of the boson. An impurity resistivity  $\rho_{imp} = 36\mu\Omega\text{ cm}$  is added. The inset shows a close-up view of the low temperature region. There is a clear deviation between  $\rho_{ph}(T) \sim \exp[-\theta_D/T]$  and  $\rho_{exp}(T) \sim T^{4/3}$  [9].

The phonon model has a critical defect at high temperatures with any reasonable parameters. While the experimental  $\rho_{exp}(T)$  show a clear saturation behavior for  $T > 160\text{ K}$ , phonon scattering results in an ever increasing T-linear resistivity for temperatures beyond a fraction of  $\theta_D$ . This saturation behavior in resistivity is a long standing problem in A-15 compounds<sup>11</sup>, transition metals, and some heavy fermion compounds such as UPt<sup>3</sup> [3]. While there is no general mechanism to explain this phenomena<sup>12</sup>, we can make the following remark.

With the spin-fluctuation model, this saturation behavior is naturally explained by the temperature dependence of the magnetic correlation length  $\xi^{-2}(T) \sim I(T)$  (see Fig.2). On the other hand, phonon scattering needs to invoke a separate mechanism to explain the saturation behavior. Recently, Calandra and Gunnarsson<sup>13</sup>, assuming lattice vibrations couple with the electron hopping integral (HI), have shown that the saturation resistivity arises at high temperatures due to a cancellation between an increasing phonon population and an increasing electron kinetic energy. Without a specific model and numerical calculations, this saturation behavior can be described phenomenologically in a two parallel resistor model ("shunting model") as<sup>14</sup>

$$\rho^{-1}(T) = \rho_{ideal}^{-1}(T) + \rho_{max}^{-1}, \quad (6)$$

where  $\rho_{ideal}(T) = \rho_{e-ph}(T) + \rho_{imp}$ , and  $\rho_{e-ph}(T)$  is calculated by Eq.(5).  $\rho_{max}$  is the maximum resistivity determined by the  $f$ -sum rule<sup>13</sup>, but here determined by empirical fitting.

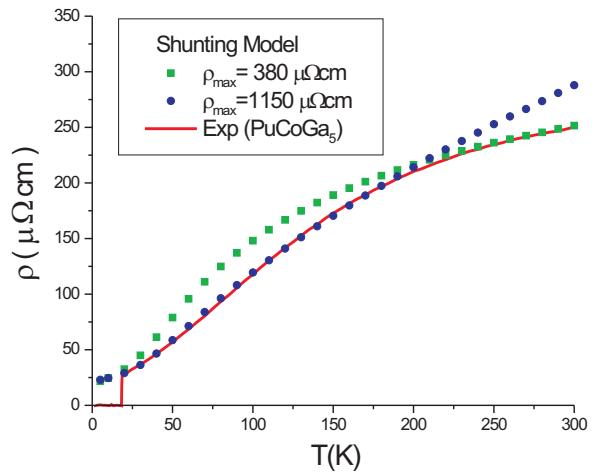


FIG. 5. Shunting model fits to the experimental resistivity (red sold line). The low temperature fit (open blue circles) is with  $\tilde{v}_{exp} = 4.9 \times 10^6\text{cm/sec}$  and  $\rho_{max} = 850\mu\Omega\text{ cm}$ . The high temperature fit (open green squares) is with  $\tilde{v}_{exp} = 3.6 \times 10^6\text{cm/sec}$  and  $\rho_{max} = 380\mu\Omega\text{ cm}$ .

In Fig.5, we show the best fit results with this model. For the electron-phonon resistivity  $\rho_{ph}(T)$ , we use  $Z=2$  and  $\theta_D = 150\text{K}$  as in Fig.4 and the overall magnitude is

tuned with  $\tilde{v}_{exp}$ . As seen, the simple shunting model cannot fit the whole temperature region with any parameters in contrast to the successful cases of A-15 compounds<sup>14</sup>. The implication is either that the shunting model is not good enough to fit the fine details or that phonon scattering is simply not the correct model for the resistivity of PuCoGa<sub>5</sub>.

### C. $T_c$

Before we estimate  $T_c$ , we need to consider the effect of impurities in the sample. The effect of impurities on  $T_c$  depends on the gap symmetry. For an s-wave gap – in the case of phonon pairing –  $T_c$  is not reduced by nonmagnetic impurities<sup>15</sup> in the first approximation. However, for an unconventional non s-wave gap – in the case of spin fluctuation pairing –  $T_c$  is reduced by any type of impurities. As noticed in the previous section, the experimental resistivity has a large residual value of  $\rho_{imp} \sim 20\mu\Omega cm$ , which should be a serious pair breaker for non s-wave pairing.

Assuming  $\rho_{imp} = 20\mu\Omega cm$  in the spin fluctuation model and plugging  $Z=4.6$  and  $\tilde{v}_{exp} = 4.78 \times 10^6 cm/sec$  in Eq.(5), we obtain  $Im\Sigma_{imp} = \Gamma_{imp} = 116 K$ . This scattering rate is far larger than  $T_c=18.5 K$ . From the theory of Abrikosov-Gor'kov<sup>16</sup>, the transition temperature is reduced by  $\sim \frac{\pi\Gamma_{imp}}{4}$ . But the original Abrikosov-Gor'kov formula should be modified by the mass renormalization factor  $Z$  as  $\frac{\pi\Gamma_{imp}/Z}{4}$ . With  $Z=4.6$  and  $\Delta T_c = T_{c0} - T_c \sim 20 K$ , we estimate the transition temperature of an ideally pure sample to be  $T_{c0} \simeq 39 K$ . Therefore, it appears that if the gap symmetry of PuCoGa<sub>5</sub> is unconventional, as in most of heavy fermion superconductors, PuCoGa<sub>5</sub> is another true high temperature superconductor. The important question is then whether the parameters of the spin fluctuation model obtained in the previous section can produce a transition temperature of  $\sim 39 K$ . For a ballpark estimate, we use the Allen-Dynes formula<sup>6</sup> for  $T_c$ ,

$$T_c = \frac{\langle \omega \rangle}{1.20} \exp\left[-\frac{1.04(1+\lambda)}{\lambda}\right], \quad (7)$$

assuming  $\mu^* = 0$  for non s-wave pairing channel.

The important parameter is  $\langle \omega \rangle = [\int_0^\infty d\omega \alpha^2 B(\omega)] / [\int_0^\infty d\omega \alpha^2 B(\omega)/\omega]$ , which defines the characteristic energy scale of the pairing boson. The nominal characteristic energy scale of spin fluctuations is  $\omega_{sf} = 150 K$ , but the above Allen-Dynes definition of  $\langle \omega \rangle$  would give several times  $\omega_{sf}$  because of the  $1/\omega$  long tail of the spin fluctuation spectra  $B(\omega)$ . In reality, the effective cut off energy scale should be between these values, say,  $3 \sim 4 \omega_{sf}$ <sup>8</sup>. Without solving the strong coupling  $T_c$  equation with details of the band structure and full dynamics of the spin fluctuations, we cannot tell the precise value of this. With this reservation for

$\langle \omega \rangle$ , the Allen-Dynes formula with  $\lambda = Z - 1 = 3.6$  and  $\langle \omega \rangle \sim \omega_{sf} = 150 K$  indeed produces  $T_c \sim 35 K$ .

For the phonon case, nonmagnetic potential scatterers do not affect  $T_c$  for an s-wave symmetry gap<sup>15</sup>. Therefore, we do not need to consider impurity effects on  $T_c$ . The Allen-Dynes formula with  $\lambda = 1$ , and  $\mu = 0.1$  gives  $T_c = 10.44$  and  $16.7 K$  for  $\langle \omega \rangle = 150 K$  and  $240 K$ , respectively. However, if we assume that the total mass renormalization ( $Z=4.6$ ) is caused solely by phonon scattering, the effective coupling  $\lambda \sim 3.6$  should be used, and it would be strong enough to produce  $T_c > 20 K$  with  $\langle \omega \rangle = 150 K$ .

### D. Natural radiation damage

PuCoGa<sub>5</sub> should have radiation-induced self damage as a function of time. A preliminary measurement<sup>1</sup> indicates a decrease of  $T_c$  at a rate of  $\sim 0.2 K/month$ , due to a radiation damage, which appears to be a quite slow suppression rate at a first look. Here, we estimate a theoretical  $T_c$  suppression with radiation damage and compare it with experiments. Because there is no study of radiation damage in PuCoGa<sub>5</sub> itself, we use results from Pu metal ( $\delta$ -phase)<sup>17</sup>. Each Pu decays into U and an  $\alpha$  particle. Both particles collide with nearby Pu nuclei creating so-called Frenkel pairs consisting of a vacancy and a self-interstitial of Pu ions. In the case of  $\delta$ -Pu, the total number of Frenkel displacements from one Pu is 0.1033 per year. Given these data, the estimated number of displacements per Pu per month is  $\sim 0.86 \%$ . Assuming the unitary limit of the impurity scattering strength, the impurity scattering rate is given by  $\Gamma_{imp} = \frac{n_{imp}}{\pi N(0)}$ , and therefore  $\Gamma_{imp} \sim 2 K$ . For s-wave pairing, this little potential scattering should have no effect on  $T_c$  suppression. However, it is not certain whether the displaced Pu ions would behave as potential scatterers or magnetic scatterers. For non s-wave unconventional pairing,  $T_c$  decreases even with potential scatterers. With a modified Abrikosov-Gor'kov formula,  $\Delta T_c = T_{c0} - T_c = \frac{\pi\Gamma_{imp}/Z}{4}$ , we obtain  $\Delta T_c \sim 0.3 K$  per month, assuming the mass renormalization factor  $Z = 5$ , consistent with the experiment. Therefore, we conclude that the  $T_c$  degradation with natural radiation damage is consistent both with unconventional pairing and also with s-wave pairing, if the displaced Pu ions behave as magnetic impurities in the latter case.

### IV. CONCLUSION:

We have found rather specific constraints on possible pairing bosons to be consistent with available experimental data for PuCoGa<sub>5</sub>. We examined two possible bosons as a common source for the normal state resistivity and the superconducting pairing.

For a spin fluctuation model, we found that the characteristic spin fluctuation energy of 150 K is consistent with the resistivity data. Theoretical calculations of resistivity produce a satisfactory agreement with the experimental resistivity: both for the anomalous power law ( $\sim T^{4/3}$ ) at low temperatures and for the saturation behavior at high temperatures. The same spin fluctuations would produce unconventional superconducting pairing, such as d-wave symmetry. In this case, the large residual resistivity  $\rho_{imp} \simeq 20\mu\Omega cm$  acts as a serious pair breaker. To survive with  $T_c=18.5$  K after this pair breaking effect, the original  $T_{c0}$  should be  $\sim 39$  K. The Allen-Dynes  $T_c$  formula can support this high  $T_{c0}$  with  $\langle \omega \rangle \sim \omega_{sf}$ . If this is the case, we have observed another unconventional high temperature superconductor after the cuprate superconductors. Perhaps this large impurity effect explains why UCoGa<sub>5</sub> ( $\rho_{imp} \simeq 20\mu\Omega cm$ ) is not a superconductor<sup>18</sup>. Taken together, we think that spin fluctuations are the most likely source of superconducting pairing as well as the normal state resistivity in PuCoGa<sub>5</sub>.

For the phonon model, we found that the most consistent phonon frequency to fit the resistivity data should be also 150 K, having a discrepancy with the estimate from the specific heat measurement ( $\theta_D = 240$  K). However, theoretical calculations of the resistivity with phonon scattering have crucial defects to explain the experimental resistivity. First, it is impossible to produce  $\rho_{exp}(T) \sim T^{4/3}$  with phonon scattering at low temperatures. Second, the saturation behavior at high temperatures needs a separate explanation – this high temperature feature in resistivity has no direct relation with the superconducting pairing mechanism, though. Putting aside these defects and pushing the possibility of the phonon pairing, the merit of phonon mediated s-wave pairing is its insensitivity to large impurity scattering observed in the sample. Using a typical strong coupling constant value  $\lambda = 1$ , it is difficult to achieve  $T_c$  of 20 K. But, assuming that phonon scattering is the main source of the large mass renormalization of  $Z \sim 4.6$ , the Allen-Dynes  $T_c$  formula with  $\langle \omega \rangle = 150$  K can easily support  $T_c$  of  $\sim 20$  K. In this case we find some similarity with the phonon mediated A-15 compound superconductors, such as Nb<sub>3</sub>Sn. However, comparing with the spin fluctuation model, we think that the phonon model is unlikely in PuCoGa<sub>5</sub>. Some key questions to be answered for the phonon model are: (1) the experimental  $\rho(T) \sim T^{4/3}$  for  $T_c < T < 50$  K [<sup>1</sup>] is not easily reconciled with a phonon scattering mechanism; (2) the absence of superconductivity in UCoGa<sub>5</sub> also needs an explanation if phonons are the pairing boson.

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$I_0 > 250K$ .